

# Aerosol Generation by Condensation in Laminar Boundary Layers for Inhalers

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## Abstract

Inhalation devices using thermal aerosol generation offer both electronic control options and cost-effective mass production. Evaporation of liquids, and subsequent dilution of the vapour with ambient air, leads to nucleation and droplet growth depending on the conditions in the mixing zone. In order to provide optimal deposition to the airways, it is important to understand the impact of both flow and design parameters on the particle size distribution. The paper describes experimental observations of aerosol generation in laminar boundary layers over a flat heated surface, wetted with a pure compound. A numerical modelling approach is also described, combining classical nucleation theory, quasi-stationary diffusion for droplet growth, and a simplified aerosol description using a single representative size parameter per cell. Compared with experiments, the model shows the correct order of magnitude of droplet sizes and a similar behaviour regarding changes of flow parameters. It also shows the striped aerosol patterns found in related experiments. A multicomponent droplet growth model is implemented in ANSYS Fluent and used to study the hygroscopic growth of droplets in the human upper airway.

## Keywords

thermal aerosol generation; boundary layer; nucleation; droplet growth

## Introduction

Evaporation-condensation inhalers evaporate a liquid drawn from a tank through a porous medium. The supersaturated vapour nucleates droplets, which grow by condensation into the mature droplets which are inhaled by the user. Such devices offer several advantages over powder and spray devices for the delivery of therapeutic compounds to the lung.

A typical cost-effective design employs a microcontroller driving a small electric heater in a flow channel. This allows precise and fast control of the heater temperature, user-friendly handling, and easy monitoring and control. Stable evaporation and flow processes are prerequisites for a narrow particle size distribution. Common carrier liquids are a mixture of 1,2-propanediol, glycerol and small amounts of water, to which is added the therapeutic compound.

The nucleation and growth is a multi-component problem. Flow and geometric parameters have an impact on particle size distribution; preceding experiments show that greater air velocities tend to produce smaller droplets, and that droplet nucleation is confined to narrow regions. Droplets begin life as a molecular cluster of the smallest size that is energetically favourable to remain condensed (nucleation) [1, 2]. A model of diffusional growth of droplets was described by [3] and developed to include mass convection by [4]. Such models have been implemented into CFD simulations of flow in a 90 degree bend [5] and in the human airway [6, 7], and have been used to explore the effect of heater power on droplet size [8].

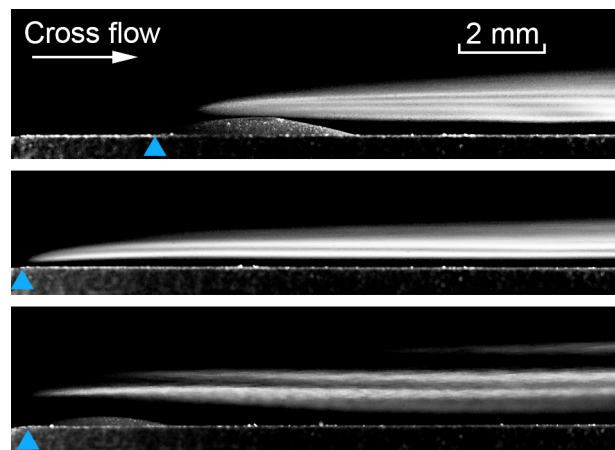


Figure 1. Instantaneous photographs of aerosol layers developing above a hot surface (200°C) in cross flow (air 20°C); wetted with 1,3-propanediol (blue triangle indicating upstream edge of wetted area, heated area begins at left image border). Top: cross flow 1.6 m/s, exposure time 400 μs; centre: 1.2 m/s, 500 μs; bottom: 0.7 m/s, 500 μs

Homogeneous nucleation is the dominant mechanism, and generates dense aerosol in a small volume. The droplets originate from supersaturated vapour without the need for existing particles acting as condensation seeds. In natural processes like cloud formation, the observed saturation ratio is often too low to allow relevant homogeneous nucleation rates, and existing particles act as condensation seeds instead. However, a vapour phase containing components with high boiling point temperatures can generate very high supersaturation leading to correspondingly high theoretical homogeneous nucleation rates.

This paper describes imaging carried out to understand the spatial distribution of condensation in idealized geometries. Spatially periodic (striped) regions of nucleation were observed. It is proposed that these are caused by the interaction of diffusion and advection of heat and mass. A first-approach model with a single condensable species was developed to explore this phenomenon. Separately, a multi-component droplet growth model was developed and integrated into a CFD solver (ANSYS Fluent) to model the growth of the droplets in the device, and subsequently in the humid environment of the lung.

## Experiments and Imaging

Three different types of imaging experiments were carried out in order to investigate aerosol generation:

1. Laminar flow of air across a plane heated surface with wetted areas (see figure 1). Cross flow was generated by a liquid piston device via a tube with nozzle outlet aligned at the upstream edge of the heated surface. A liquid patch was deposited manually on the surface.

2. A jet-in-cross-flow (JICF) setup with a smooth round vapour jet placed at a right-angle to the outlet flow of a small open wind tunnel which was operated in laminar conditions (see figure 2). The vapour jet was generated by a closed evaporator. Cross flow velocity was directly measured with a hot-wire anemometer, while jet velocity was estimated by particle image velocimetry of the fastest upward motion.
3. A falling hot droplet in stationary air. Liquid was slowly fed through a heated vertical pipe with an sharp edged outlet of 2 mm diameter (see figure 3). Photographs were taken at different heights below the outlet, and fall velocity was measured by image velocimetry.

The high-speed camera was a Vision Research PHANTOM v1210 monochrome equipped with a 65 mm 1–5x macro lens. Lighting used either an LED spotlight or a pulsed laser with light sheet optics.

Figure 1 shows typical stratified aerosol layers, the corresponding video recordings showed a steady-state flow without any oscillations (except for transition to turbulent flow further downstream).

Figures 2 and 3 exhibit spatially periodic (striped) regions of nucleation. In JICF setups (figure 2), the periodic nucleation was stable in time. The frequency  $f$  was strongly correlated to the cross flow velocity  $u$ , exhibiting a dependency of roughly

$$f \sim u^{1.5} \quad \text{for } 0.5 \text{ m/s} \leq u \leq 1.2 \text{ m/s} \quad (1)$$

while not showing any synchronicity with vortices developing downstream of the jet.

In the falling droplet experiments (figure 3) one could see the striped patterns developing near the stagnation point and then being carried downstream.

Additional measurement of droplet size distributions were carried out with 1,3-propanediol being evaporated from a flat porous heater chip of 2 mm × 2 mm in a closed housing with cross airflow in the range of approx. 0.4 to 4 m/s. Median values were 1.5 to 2.5  $\mu\text{m}$ , 10th percentiles were 0.7 to 0.9  $\mu\text{m}$ , and 90th percentiles were 3 to 6  $\mu\text{m}$  (mass based), showing decreasing sizes with increasing velocities. Often bimodal distributions were found.

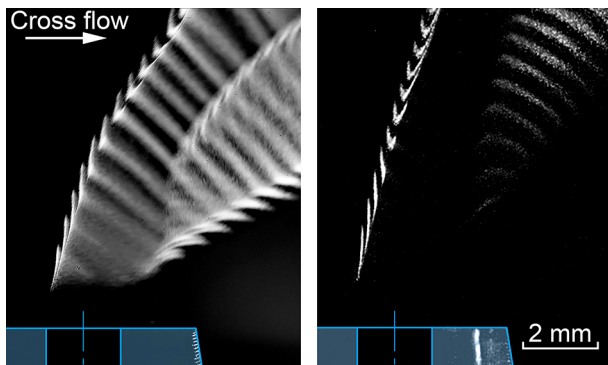


Figure 2. Instantaneous photographs of smooth round jet in cross flow with striped aerosol patterns; jet: 1,2-propanediol at 188 °C, cross flow: air at 20 °C. Left: Illumination with lateral spotlight, cross flow 0.7 m/s, jet velocity 1.1 m/s, exposure time 98  $\mu\text{s}$ . Right: Laser light sheet illumination in centre plane of jet parallel to image plane, cross flow 0.8 m/s, jet 1.4 m/s, laser pulse duration 0.2  $\mu\text{s}$ . Nozzle contour added.



Figure 3. Instantaneous photograph of striped pattern of aerosol generation around a falling hot droplet in air (1,2-propanediol / glycerol mixture with mass fractions 2:1, diameter 2.85 mm, droplet temperature 180 °C, fall velocity 0.41 m/s, air 20 °C, exposure time 120  $\mu\text{s}$ )

## Boundary Layer Model of Spatially Periodic Nucleation

### Model Scheme

Preliminary experiments showed little impact of liquid composition within the ranges usually used in devices of this type. In order to reduce complexity, a single condensable component modelling approach was chosen, substituting 1,3-propanediol for mixtures of 1,2-propanediol and glycerol as it has thermal properties in between. The model is non-steady, and it uses:

- an ideal mixture gas phase consisting of dry air and vapour of the single condensable component.
- a vastly simplified flow field, obtained by enforcing a one-dimensional laminar boundary layer profile upon the whole simulation region. Advection was modelled by shifting calculation cells in the streamwise direction, avoiding numerical diffusion effects on the droplet population caused by advective inter-cell transport.
- a concentration field operated on by advection (implicitly by shifting cells), diffusion, nucleation and droplet growth of the condensable species. An explicit finite volume approach for mass and thermal diffusion was used.
- a homogeneous nucleation rate  $J$  according to one-component classical nucleation theory [2], with droplets being initiated at radii slightly larger than the critical radius, allowing them to grow initially. A threshold was applied to the droplet number, obtained from integration of  $J$ , to define the event of the first nucleation and start of population growth in a cell.
- a droplet growth model based on diffusional mass and heat transfer in a quasi-steady-state spherically symmetric system of a droplet in an infinite far field (Maxwell [3]). In each time step the droplet temperature was set to the equilibrium state of mass and heat transfer.
- representation of the aerosol population in each cell using a single diameter. Accompanying simulations with multiple diameter classes showed that the mean-mass diameter is—in this case—advantageous over other characteristic diameters, e. g. the Sauter mean diameter.

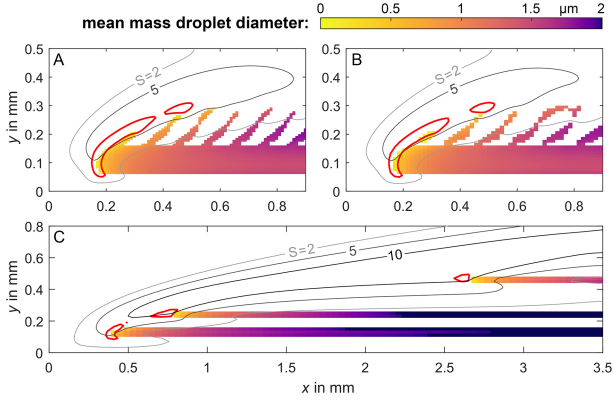


Figure 4. Simulation results for nucleation in boundary layers. **A:** 1,2-propanediol with surface temperature of 188°C and cross flow velocity of 0.7 m/s (constant), wetted area begins at  $x = 0$  mm, heated area at  $x = -0.3$  mm. **B:** same as A, coarser grid. **C:** 1,3-propanediol with 200°C, 1.2 m/s (const.), wetted area begins at  $x = 0$  mm, heated area at  $x = -0.5$  mm. Red lines: nucleation rate  $J = 5 \cdot 10^8 \text{ mm}^{-3} \text{ s}^{-1}$ , grey lines: saturation ratio  $S$ , droplet size is colormapped.

- a 2-dimensional rectangular uniform grid of up to  $250 \times 100$  cells with 4 state variables per cell (gas phase temperature, concentration of condensable component, number of droplets, quantity of condensed matter).
- properties of pure components, including vapour pressure curves, taken from [9, 10] and estimation of mixture properties of vapour and air as described in [11].

The model was implemented in MATLAB, defining a set of explicit ordinary differential equations (ODE) to be solved over time. It has to be stated that the model—as it is implemented—violates the state equation of the ideal gas in order to impose a given isobaric condition *and* equimolar diffusion on a uniform rectangular grid. This was accepted as the purpose of the model was to gain a basic understanding of observed aerosol pattern phenomena.

### Simulation Results

Boundary conditions for the simulations of heated wet planes in cross flow were: air entering at west inlet, south boundary a wall with fixed temperature and concentration in sections, north boundary adiabatic and diffusion barrier.

Simulation results of boundary layers over heated planes exhibit key similarities to the patterns found in experiments, see figure 4. Stratified multiple layers were found in the majority of simulations, with location and spacing mainly influenced by the thickness of the thermal and concentration boundary layers as well as the cross flow velocity.

With lower velocities, the simulations also showed spatially periodic patterns, resembling those of JICF experiments. In the heated plane experiments the periodic patterns could only be observed with slightly inclined cross flow (not shown here).

Droplet numbers leaving the simulation region at the east outlet were used to estimate final droplet sizes under equilibrium conditions in the aerosol, showing the same order of magnitude as the experiments. The simulated distributions were narrower and less smooth. The impact of cross flow velocity on the median diameter was stronger than in the experiments. Sizes varied between the different stratified layers, delivering a possible explanation for measured bimodal distributions.

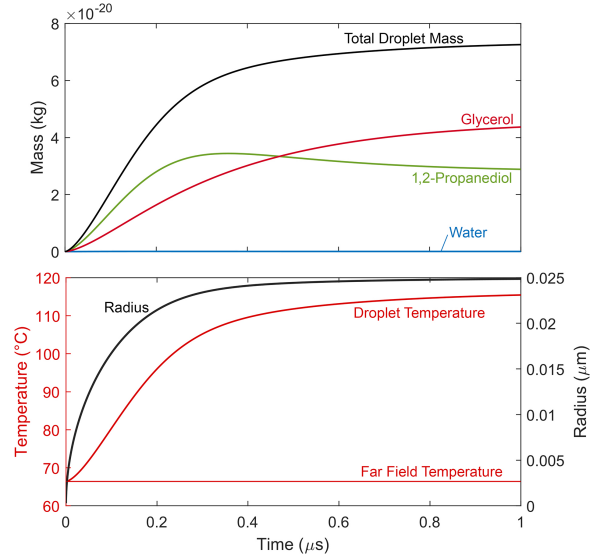


Figure 5. Growth of a droplet in a mixture of water, 1,2-propanediol and glycerol. Ambient vapour conditions were set as 0.7% water, 8.8% 1,2-propanediol, 4.2% glycerol and 86.3% air (mass fractions) at 66.4°C.

## Multiple Condensable Component Model of Droplet Growth

### Model Scheme

Liquids used in the context of this paper are often mixtures, and aerosol laden flow travels through airways with water bearing surfaces during inhalation. Aerosol droplets are therefore expected to change composition and size, depending on the conditions of the surrounding vapour/gas-phase.

In order to investigate the behaviour of droplets with multiple condensable components plus air as a non-condensing gas, a second model was set up. It describes the growth of a single droplet in a saturated or supersaturated gaseous environment containing multiple condensable vapour components *after* nucleation.

In the growth process the diffusional mass transfer of a single component  $i$  is represented by

$$\frac{dm_i}{dt} = 4\pi r D_i (\rho_i - \rho_{r,i}) \quad (2)$$

where  $m_i$  is the mass of component  $i$ ,  $r$  is the droplet radius,  $m = \sum m_i$  is droplet total mass,  $D_i$  is the diffusion coefficient of component  $i$ ,  $\rho_i$  and  $\rho_{r,i}$  are vapour partial densities of component  $i$  in the far field and at the droplet surface.

The change in temperature of the droplet is calculated by

$$\frac{dT_r}{dt} = \frac{1}{m c_{p,\text{mix}}} \left( L_{\text{mix}} \frac{dm}{dt} + 4\pi r K (T - T_r) \right) \quad (3)$$

which accounts for the effects of latent heat of vapourisation and conduction.  $T_r$  is the droplet temperature,  $c_{p,\text{mix}}$  is the specific heat of the droplet mixture,  $L_{\text{mix}}$  is the latent heat of the droplet mixture,  $K$  is the thermal conductivity of the surrounding vapour and  $T$  is the far field temperature.

Properties of pure components were taken from [9, 10]. Values for  $D_i$  were estimated as binary diffusion coefficients in air using the Fuller method [12]. Mixture properties were calculated applying methods from [11]. Vapour-liquid-equilibrium of mixtures were estimated with a modified UNIFAC-method [13, 14].

It was assumed that the initial composition is solely made up of the component with the lowest saturation pressure, and that the initial droplet size is slightly larger than its critical radius, allowing the droplet to grow from its otherwise equilibrium initial state. The set of differential equations (equations 2 and 3) was solved using the ode15s function in MATLAB and also coded into a user defined function (UDF) for further application in ANSYS Fluent.

### Simulation Results

Figure 5 shows the growth of a droplet in a mixture of water, 1,2-propanediol and glycerol from near the critical radius. This simulation time of  $1\ \mu\text{s}$  was chosen to demonstrate the stiff initial behaviour of the multicomponent growth model. The volume in which the droplet grows was limited to  $1\ \mu\text{m}^3$ , so the effect of growth stagnation due to depletion of the vapour can be observed.

Figure 6 shows simulation results of droplets growing due to hygroscopic growth in a MRI scan of a human upper airway. The initial diameter was chosen after using the multicomponent model UDF in simulations of an inhaler device, which produced an average droplet diameter of  $0.7\ \mu\text{m}$ . The composition of these droplets was then initialised at the inlet to the airway simulation and flow rate was set to  $1.1\ \text{l/min}$ . The droplets grow from  $0.7$  to in excess of  $2\ \mu\text{m}$ . This growth is necessary to ensure the droplets deposit in the deep lung, without being exhaled.

### Conclusions

Two models addressing aspects of nucleation and droplet growth were developed. The first model is a single component model which simulates the interaction of mass and heat transport in combination with nucleation and condensation. It was used to investigate the local aerosol generation process in the evaporation zone of the device and to estimate impacts of flow and design parameters. Despite its simplifications, it delivers similar results to experiments and possible explanations for spatial condensation patterns and bimodal size distributions. The

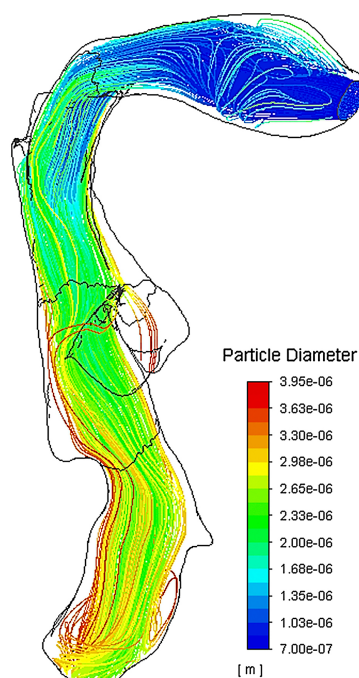


Figure 6. Simulation of droplets growing due to hygroscopic growth in the upper human airway (adult male).

second model describes droplet growth in multiple condensable component systems. It was coded as a user defined function in ANSYS Fluent and will be used in further investigations of the impact of device geometry and airway flow.

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